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Research Domain, discipline, and sub-discipline

Chemistry, Theoretical Chemical Physics, Time-Dependent Electronic Structure Theory

Title of Submission

Future Needs for Advanced Cyberinfrastructure to Support Quantum Dynamic Simulations and Computational Time-Resolved Spectroscopy

Abstract (maximum ~200 words).

The last two decades have witnessed a revolution in ultrafast time-resolved spectroscopy with the development of multidimensional techniques. The complex yet extremely physics-rich signals encode crucial and fundamental processes that require not only quantum dynamic simulations, but also innovative spectral analysis protocols to connect physical observables to fundamental theories. The knowledge of photo-physical processes underlying electrophotonic, photoelectronic, spintronic, and photocatalytic materials functionalities gleaned through application of quantum dynamics will facilitate the advancement of technologies that directly impact society.
There is a clear need for advanced cyberinfrastructure that seamlessly integrates modern high-performance computing concepts with time-dependent quantum mechanical theories and modular high-performance numerical libraries that are highly parallelized, extensible, and reusable.

Question 1 Research Challenge(s) (maximum ~1200 words): Describe current or emerging science or engineering research challenge(s), providing context in terms of recent research activities and standing questions in the field.

Technology for producing ultrafast high-fluence laser pulses has spawned a new generation of spectroscopies for probing linear and nonlinear optical response in matter on the very timescale that photo-physical processes occur. Multidimensional time-resolved spectroscopies using time delayed sequences of these ultrafast pulses to excite and subsequently probe the system of interest have provided unprecedented capabilities for observing ultrafast photo-induced molecular processes that underpin photoelectronic, electrophotonic, photocatalytic, and spintronic technologies.

However, the need for theoretical insights into time-resolved multidimensional spectroscopy experiments has, thus far, gone essentially

unanswered by computational physical scientists working in this field. Implicit to this challenge is the need to treat both the non-equilibrium, excited state electronic structure and the dynamical evolution of molecular degrees of freedom over the course of the modeled experiment. This treatment has to be accurate enough to provide a reliable quantum mechanically-grounded interpretation of intricate experimental spectra. Quantum dynamics will provide a time-dependent, state-specific interpretation of the chemical dynamics encoded in multidimensional spectroscopies, and aid in the design of new molecular and material systems exhibiting desired optical/magnetic characteristics, with the potential for revolutionary impact in the broader scientific community and beyond.

The grand challenge largely lies in the development of a computational platform that can model time-dependent characteristics of a quantum system and the interactions with electromagnetic fields without assuming constant quantum state populations in time. On one end of the spectrum, the computational method needs to be capable of applying time-dependent quantum theory to model excited state quantum dynamics using field parameters from experiment. On the other, the same computational platform has to be efficient to compute dynamics for different pulse sequences to resolve all signals comprising a multidimensional spectrum. However, existing computational software packages are incapable of reproducing time-resolved spectroscopic signals and high-order spectroscopic properties that are crucial to modern-day molecular and materials design. There have been emerging efforts, including those from the Li research group, to develop efficient algorithms and computational software package that are able to model time-resolved multidimensional spectroscopy with atomic resolution quantum dynamics. The success of applying quantum dynamics to model multidimensional spectroscopy hinges on not only the new development of software with capabilities described above, but also advanced computing infrastructure designed to support quantum dynamic simulations.

Question 2 Cyberinfrastructure Needed to Address the Research Challenge(s) (maximum ~1200 words): Describe any limitations or absence of existing cyberinfrastructure, and/or specific technical advancements in cyberinfrastructure (e.g. advanced computing, data infrastructure, software infrastructure, applications, networking, cybersecurity), that must be addressed to accomplish the identified research challenge(s).

Quantum dynamics is intrinsically a four-dimensional theoretical problem that propagates three-dimensional quantum wave function in time. We shall use the real-time time-dependent density functional theory (RT-TDDFT) for electronic dynamics as an example to illustrate the need for advanced cyberinfrastructure.

In the case of RT-TDDFT, while the three-dimensional electron density at each time step is associated with a spatial integration over grid, which is ideal for wide-SIMD parallel infrastructure, the time-integration of the three-dimensional electron wave function or density is strictly sequential. For example, to obtain one picosecond electronic dynamics driven by multiple laser pulses, one million sequential time-integrations of three-dimensional spatial integration of the Hamiltonian over a grid are carried out (assuming the standard one attosecond per time-integration step). As a dynamic method, errors are cumulative over time. As a result, ultrafine or superfine grid with a few hundreds of millions of grid-points are commonly used to integrate the DFT molecular Hamiltonian, especially when the external perturbation is strong and DFT with generalized gradient approximation (GGA) and meta-GGA are applied. When nuclear quantum wave functions are also considered together with electronic wave function, the computational cost grows exponentially with the system size. Such a computational framework renders quantum dynamics extremely computational resource demanding.

If the software is designed and written with high concurrency at its core for quantum dynamics, wide-SIMD computer infrastructure can be fully taken advantage of. The strictly sequential nature of quantum dynamics also calls for high-performance computing infrastructure with low latency communication.

The largest performance gain for quantum dynamics is when all fundamental integrals can be stored in memory (i.e. the so called “in-core” method) as the parallel load balance and concurrency can be highly optimized. However, electron repulsion integral is a rank-4 tensor and storing all integrals in memory is impractical (e.g. a system with 1000 basis functions will need 1TB RAM to store all integrals). As a result, electronic structure community often chooses to use the “direct” algorithm in which integrals are computed and re-computed on-demand. While this allows applications of large scale systems without the limitation of memory, this gives rise to a major slow-down in quantum electronic dynamics due to not only the redundant integral operations, but also the loss of optimized load balance and concurrency.

The complex computational framework of quantum dynamics demands that software design has high vectorization and high concurrency in its core, and computer infrastructure has wide-SIMD, low latency communication, and extremely large RAM (or even possible RAM/flash-drive hybrid storage).

Question 3 Other considerations (maximum ~1200 words, optional): Any other relevant aspects, such as organization, process, learning and workforce development, access, and sustainability, that need to be addressed; or any other issues that NSF should consider.

The success of an advanced cyberinfrastructure project on quantum dynamics hinges on a close collaboration of theoretical chemical physicists, applied mathematicians, software and hardware engineers. This requires that domain scientists have necessary background in software engineering and applied math in order to facilitate the development because without effective communication among scientists and engineers in different research domains, such a collaborative project will unlikely generate impactful products.

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